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EXAMINATION OF CHEMICAL KINETICS DATA USED IN INDUCTION ZONE CALCULATIONS FOR THE COMBUSTION OF CORDITE

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The rate constant data used by J. G. Sotter (1965) to model the induction zone of a double-base propellant are re-examined in the light of recent data. Revised constants are reported for most of the 20 reactions considered by Sotter; about five of the changes appear significant.		

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I. INTRODUCTION

In 1965 a paper appeared[1] in the Tenth Symposium (International on Combustion) by J. George Sotter of Sheffield University, Sheffield, England in which the author constructed profiles of velocity, and temperatures vs distance for the induction and explosion zones above cordite burning at 1000 psi (7 MPa). For this purpose it was necessary to select pertinent reactions and to assemble reaction rate expressions for them. The author selected a total of twenty reactions and carried out computer calculations using them.

In the present report the rate constant data used are examined in the light of current knowledge of these reactions and, where necessary, revised values are suggested.

II. SOURCES OF REACTION RATE CONSTANTS

It is an unusual, and unfortunate aspect of Sotter's paper that not only were the pertinent rate constant data not tabulated, but the reference source[2] was a paper given at a meeting in 1962 of the Western States Section of the Combustion Institute. This paper was not published later in a journal and considerable library time was spent trying to find it. A copy was finally obtained through the kindness of Ms. Rebecca Palmer, Secretary to Professor Richard Sawyer, Department of Mechanical Engineering, University of California who had presided at the particular meeting. The reactions considered by Sotter and the rate constants he used are shown in Table I.

Initially it was planned to survey the literature for the individual reactions used by Sotter and to evaluate the results of this survey. It soon became evident that this was a task of very considerable magnitude. Fortunately the examination of the literature revealed the existence of critical surveys of reaction rate data which included most of the reactions of interest. Since the surveys originated in laboratories where considerable expertise existed in connection with reactions of the type of interest here, the values reported here are for the most part taken from those surveys. In Table I the values recommended are shown along with those used by Sotter.

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[1] *J. G. Sotter, Tenth Symposium (International) on Combustion, 1405, (1965).*

[2] *G. E. Ellis and G. S. Bahn, "Engineering Selection of Reaction Rate Constants for Gaseous Chemical Species at High Temperatures", 1962 Fall Meeting, Western States Section, The Combustion Institute.*

III. REMARKS ON CURRENT CONSTANTS AND THOSE USED BY SOTTER

Of the twenty reactions cited by Sotter, only seventeen are independent. Reactions (4b), (5b), and (6b) of Table I were assigned constants on the basis of applying an arbitrary "catalyst-efficiency" factor to the rate constants for reactions (4), (5), and (6), respectively.

In considering such a large number of reactions, it is a problem deciding where to start. It is an additional problem to decide how significant discrepancies between current values and those used by Sotter are. In his paper Sotter emphasized the importance of the "water-gas equilibrium" reactions, reactions (1) and (7) of Table I and of reaction (17), the termolecular reaction between nitric oxide and hydrogen. He states, in addition, that reactions (8), (9), (10), (12), and (14) could probably be omitted without loss of accuracy.

It has therefore seemed desirable to add a few comments about these reactions. Since no new data have been found for reaction (17), comments will be made only for the other two.

Reaction (1)

$$H + H_2O \rightarrow H_2 + OH$$

has been considered by the Leeds group[3] and by Wilson[4]. The value recommended by the former for the reaction for which Sotter lists the value, i.e., the back reaction

$$OH + H_2 \rightarrow H + H_2O$$

is $k = 2.19 \times 10^{13} \exp(-5150/RT)$ $cm^3\ mole^{-1} sec^{-1}$ ±35% and is given for the temperature range 300-3000K. Wilson[4] recommends

$k = 2.3 \times 10^{13} \exp(-5200/RT)$ $cm^3 mole^{-1} sec^{-1}$ for the temperature range 300-2000K. The two values are thus in close agreement and differ somewhat from the value used by Sotter. At 1000K for example the expression used by Sotter yields a rate constant some thirty times as large as that derived from the above values.

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[3] *(a) D. L. Baulch et al., "High Temperature Reaction Rate Data", No. 1, Dept of Physical Chemistry, The University, Leeds, 2, England; May 1968 (b) ibid, No. 2; Nov 1968 (c) ibid No. 3; Apr 1969 (d) ibid, No. 4; Dec 1969 (e) ibid, No. 5; Jul 1970.*

[4] *W. E. Wilson, Jr., "A Critical Review of the Gas Phase Reaction Kinetics of the Hydroxyl Radical", J. Phys. Chem. Ref. Data, 11, 535 (1972).*

Perhaps the most serious revision of all occurs for reaction (7)

$$CO + OH \rightarrow CO_2 + H$$

The value used by Sotter was taken from the work of Westenberg and Favin[5] at the Applied Physics Laboratory,

$$k_f = 10^{13} \exp(-10{,}000/RT) \text{ cm}^3\text{mole}^{-1}\text{sec}^{-1}$$

This expression was based on measurements made in the authors' laboratory at elevated temperatures and on data reported for room temperature measurements in the Russian literature[6]. Further studies in Westenberg's laboratory[7] showed the Russian work to be in serious error. Reaction rate constants have been evaluated as shown below.

Measured Rate Constant for CO + OH

in $\text{cm}^3\text{mole}^{-1}\text{sec}^{-1}$

T(K)	k_f
298	8.0×10^{10}
523	8.7×10^{10}
915	13.1×10^{10}

The data indicate an activation energy of zero. A general expression of the following type can be written

$$k_1 = \frac{2.5 \times 10^{13}}{T\Pi[1-\exp(-h\nu_i/kT)]}$$

where five vibrations must be considered for ν_i: 1670 cm^{-1} (C-O stretch), 1300 cm^{-1} (O-H bend), 830 cm^{-1} (H out-of-plane bend). Values are given in the reference for the denominator from 300-1000K. Wilson[4] recommends for the range 300-2000K

$$k = 3.1 \times 10^{11} \exp-(300/T) \text{cm}^3\text{mole}^{-1}\text{sec}^{-1}$$

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5 *A. A. Westenberg and S. Favin, Ninth Symposium (International) on Combustion, Academic Press, 1963, p. 785.*

6 *L. I. Avramenko and R. V. Lorentso, Zhur. Fiz. Khim., 24, 207 (1950).*

7 *A. A. Westenberg and N. DeHaas, J. Chem. Phys., 58, 406 (1973).*

which yields $k = 2.3 \times 10^{11} sec^{-1}$ at 1000K. Westenberg and DeHaas report $k = 1.4 \times 10^{11} sec^{-1}$. The Leeds Group [3a] recommends

$$k = 5.6 + .8 \times 10^{11} \exp(-1080 \pm 500/RT) cm^3 mole^{-1} sec^{-1}$$

At 1000K this yields $k = 3.4 \times 10^{11} sec^{-1}$ which is also in good agreement. There appears to be little to choose from among these. I favor the data of Westenberg and DeHaas but would like to point out that the other expressions yield rather similar results.

Where the remaining rate constants show differences, they are much smaller and I am not able to evaluate their importance for the results obtained by Sotter. The groups cited seem most reliable, and since no serious differences exist for those rate constants for which values are cited independently by both of them, the recommended values should be reliable.

Note that in Table I rate constants are cited for either the forward or back reaction in the reversible reactions but not for both. Sotter obtained equilibrium constants[2] for all reactions except No. 7 of Table I. The equilibrium constants in reference 2 were calculated from data tabulated in the JANAF Interim Thermochemical Tables[8]. Equilibrium constants are generally available in more reliable form than are rate constants and, since the cited source is one whose authority is widely accepted, I have not felt it necessary or desirable to calculate and tabulate either equilibrium constants or the other rate constant for each reversible reaction. It is clear, of course, that the combination of an incorrect k_f and a correct equilibrium constant leads to an incorrect k_b.

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[8] *D. R. Stull et al, JANAF Interim Thermochemical Tables, Thermal Laboratory, Dow Chemical Co., Midland, MI, Dec 31, 1960, eq seq.*

Table I. Reaction Rates Constants Used by Sotter and Values Recommended from More Recent Compilations

(Values used by Sotter were taken from reference 2 except for Reaction (7) which was taken from reference 5, and Reaction (17) which is given by Sotter along with the reference source. For the Recommended Rate Constants, all values were taken from reference 3 except for Reaction (7), which is discussed earlier in this report, and Reaction (13) for which the source is reference 9.

No.	Reaction	Sotter's Value	Current Recommendation
(1)	$H + H_2O \rightarrow H_2 + OH$	$k_b=3\times10^{14}\exp(-2500/RT)$	$k_b=2.2\times10^{13}\exp(-5150\ \pm150/RT)$
(2)	$H + O_2 \rightarrow O + OH$	$k_f=4\times10^{14}\exp(-18000/RT)$	$k_t=2.2\times10^{14\pm.3}\exp(-16800\ \pm500/RT)$
(3)	$O + H_2 \rightarrow OH + H$	$k_f=3\times10^{14}\exp(-6000/RT)$	$k_f=1.7\times10^{13\pm.2}\exp(-9450\ \pm250/RT)$
(4)	$H_2O + M \rightarrow H + OH + M$	$k_f=10^{23}T^{-1.5}\exp(-114730/RT)$	[a] $k_f=1.6\times10^{18}\exp(-119000/RT)$
(4b)	$H + OH + NO \rightarrow H_2O + NO$	k_b for (4) multiplied by factor	no new data
(5)	$H_2 + M \rightarrow 2H + M$	$k_f=10^{21}T^{-1.5}\exp(-103240/RT)$	no new data
(5b)	$H + H + NO \rightarrow H_2 + NO$	k_b for (5) multiplied by factor	no new data
(6)	$O_2 + M \rightarrow 2O + M$	$k_f=1.1369\times10^{25}T^{-2.5}\exp(-118000/RT)$	no new data
(6b)	$O + O + NO \rightarrow O_2 + NO$	k_b for (6) multiplied by factor	no new data
(7)	$CO + OH \rightarrow CO_2 + H$	$k_f=10^{13}\exp(-10000/RT)$	[b] $k_k=\frac{2.5\times10^{13}}{T\pi[1-\exp(-h\nu_i/kT)]}$ $k_f=5.6\pm.8\times10^{11}\exp(-1080\pm500/RT)$
(8)	$NO + M \rightarrow N + O + M$	$k_f=5.18\times10^{21}T^{-1.5}\exp(-150000/RT)$	$k_f=1.1\times10^{21}T^{-1.5}\exp(-150000/RT)$
(9)	$N_2 + O \rightarrow NO + N$	$k_f=5.5\times10^{11}T^{0.5}\exp(-72000/RT)$	[c] $k_f=1.36\times10^{14}\exp(-75000/RT)$

No.	Reaction	Sotter's Value	Current Recommendation
(10)	$NO + O \rightarrow O_2 + N$	$k_f=3.2\times10^{9}\exp(-39100/RT)$	$k_f=1.5\times10^{9}T\exp(-38640/RT)$
(11)	$N_2 + O_2 \rightarrow 2NO$	$k_f=9.1\times10^{24}T^{-2.5}\exp(-128500/RT)$	no new data
(12)	$O + N_2O \rightarrow 2NO$	$k_b=1.0\times10^{11}\exp(-15500/RT)$	$k_b=2.3\times10^{13}\exp(-24100/RT)$
(13)	$N_2O + M \rightarrow N_2 + O + M$	$k_f=2.07\times10^{22}T^{-2}\exp(-62000/RT)$	$k_f=4.88\times10^{14}\exp(-58000/RT)$
(14)	$N_2 + O_2 \rightarrow N_2O + O$	$k_f=4.0\times10^{9}\exp(-93200/RT)$	no new data
(15)	$O + H_2O \rightarrow 2OH$	$k_b=3\times10^{14}\exp(-2500/RT)$	$k_b=5.8\times10^{12\pm.45}\exp(-780\pm500/RT)$
(16)	$OH + M \rightarrow H + O + M$	$k_f=10^{21}T^{-1.5}\exp(-101000/RT)$	no new data
(17)	$2NO + H_2 \rightarrow 2HNO$ ($+H_2 \rightarrow 2H_2O + N_2$)	$k_f=5\times10^{18}\exp(-47000/RT)$	no new data

[a] Estimated from an estimate in reference 3 that E = 119,000 and that at 200K, $k=3.4\times10^{5}cm^{3}mole^{-1}sec^{-1}$

[b] ν=cf. remarks in text

[c] Accuracy given as ±100%

[9] *K. Schofield, "Evaluated Chemical Kinetic Rate Constants for Various Gas Phase Reactions", J. Phys. Chem. Ref. Data., 2, 25 (1973).*

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9. K. Schofield, "Evaluated Chemical Kinetic Rate Constants for Various Gas Phase Reactions," J. Phys. Chem. Ref. Data., 2, 25 (1973).

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